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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF	:	
KENICHI MOTOYAMA, ET AL	:	GROUP: 1796
SERIAL NO: 10/583,557	:	
FILED: JUNE 19, 2006	:	EXAMINER: LOEWE, R.
FOR: WATER REPELLANT COATING FILM HAVING LOW REFRACTIVE INDEX	:	

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

The following is an appeal to the Board of Appeals concerning the decision by the Examiner to continue the rejection of Claims 1-6 and 16-25 of the above-identified application as follows:

REAL PARTY OF INTEREST

Nissan Chemical Industries, Ltd. is the real party of interest in the above-identified application.

RELATED APPEALS AND INTERFERENCES

There are no applications on appeal or in interference proceedings of related cases at the Board of Appeals and Interferences.

STATUS OF CLAIMS ON APPEAL

Claims 1-6 and 16-25 are pending and are on appeal in the application. Claims 7-15 have been canceled.

STATUS OF AMENDMENTS

No amendment has been filed in response to the Office Action of January 14, 2009 which is a third action on the merits of the case.

SUMMARY OF CLAIMED SUBJECT MATTER

The present invention is directed to a process for preparing a coating fluid containing a polysiloxane, as set forth in Claim 1, by:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$Si(OR)_4$,

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3$ (2)

wherein R^1 is a C_{1-5} alkyl group, and n is an integer ranging from 0 to 12,

a silicon compound (C) of formula (3):

$H_2NCONH(CH_2)_mSi(OR^2)_3$ (3)

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

R^3CH_2OH (4)

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from

the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group, and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 ,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranging from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranging from 0.5 to 100 mol and the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranging from 0.2 to 2 mol, and

heating this reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water.

Support for the invention as claimed may be found in the paragraph bridging pages 6 and 7 of the text of the application.

Another aspect of the invention is claimed in Claim 4 and is directed to a process for forming a coating film containing a polysiloxane by:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$Si(OR)_4$,

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer ranging from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group, and

oxalic acid (E),

wherein

(iv) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 ,

(v) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranging from 0.01 to 0.20 mol,

(vi) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranging from 0.5 to 100 mol and the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranging from 0.2 to 2 mol, and

heating this reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water.

Support for the invention as claimed may be found in the paragraph bridging pages 6 and 7 of the text of the application.

Another aspect of the invention is claimed in Claim 17 and is directed toward having a refractive index ranging from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, which is formed as adhered to a substrate surface by:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer of from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups present in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol and the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from of 0.2 to 2 mol;

heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt, as calculated from silicon atoms in the reaction mixture and while in the absence of water, and

forming a solution of a polysiloxane, and

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating, and

heat-curing the coating at a temperature ranging from 40 to 450° C.

Support for the invention as claimed may be found in the paragraph bridging pages 6 and 7 of the text of the application and in the preamble of original Claim 7.

Still another aspect of the invention is claimed in Claim 20 and is directed to a process for forming a coating film by:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a $C_{1\text{-}5}$ alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a $C_{1\text{-}5}$ alkyl group, and n is an integer ranging from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol, and

(iv) the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.2 to 2 mol;

heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water forming a solution of a polysiloxane; and

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating;

drying the coating at a temperature ranging from 40 to 150° C, and

aging the coating at a temperature of from 20 to 100° C for curing, to form a coating film having a refractive index of from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, as adhered to the substrate surface.

Support for the invention as claimed may be found in the paragraph bridging pages 6 and 7 of the text of the application and in the preamble of original Claim 7 and in the text on page 18, lines 7-14.

Another aspect of the invention is claimed in Claim 23 and is directed to a process for forming a coating film having a refractive index of from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, which is formed as adhered to a substrate surface by:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a $C_{1\text{-}5}$ alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a $C_{1\text{-}5}$ alkyl group, and n is an integer ranging from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a $C_{1\text{-}5}$ alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol, and

(iv) the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.2 to 2 mol;

heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water forming a solution of a polysiloxane; and

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating;

drying the coating at a temperature ranging from 40 to 150° C, and

aging the coating at a temperature of from 20 to 100° C for curing, to form a coating film.

Support for the invention as claimed may be found in the paragraph bridging pages 6 and 7 of the text of the application and in the preamble of original Claim 7 and in the text on page 18, lines 7-14.

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Whether Claims 1-6 and 16-25 stand properly rejected based on 35 USC 103(a) as unpatentable over Nogami et al, U.S. Patent 5,800,926 in view of Hayashi et al, U.S. Patent 6,800,330.

ARGUMENT

Claim Rejection, 35 USC 103; Rejection of Claims 1-6 and 16-25

The Examiner summarizes on page 5, first full paragraph of the Office Action of January 14, 2009 that the combination of Nogami et al and Hayashi et al *collectively teaches a process for preparing a coating fluid according to instant Claims 1-3, a process for forming a coating film, a coating film, and other invention aspects*. However, appellants disagree with this view of the art relative to the two cited references quite substantially. It first must be established that the process of Nogami et al is of the same type or similar to that of the present invention insofar as it teaches the reaction of a mixture prepared from a tetraalkoxysilane, the silicon compound (B) of formula (2) and alcohol (C) *in the absence of water at a temperature of 50 to 180 ° C to form a polysiloxane solution*. Appellants emphasize that both the present invention and the process of the reference occurs in an anhydrous medium (no water present). On the other hand, Hayashi et al discloses a process of producing a coating or a film which is essentially silica based (see column 1) by preparing a mixture of at least one of the compounds (1), (2) and (3) in the presence of a basic catalyst and water, which is in direct contradiction of the teachings of Nogami et al. Thus, the

reaction medium of Hayashi et al is not anhydrous, and the product obtained ((A) component) by hydrolysis and condensation is substantially silica. (Hydrolysis produces a silica material containing a substantial amount of silanol groups, many of which react with each other with the elimination of water to form Si-O-Si bonds.) (The term "condensation" means a condensate in which a slight proportion of the silanol groups have been condensed and a mixture of condensates which differ from each other in the degree of condensation (see column 2, lines 55-59).) Clearly, the first stage reaction material (A) of the Hayashi et al patent is not at all produced by the process disclosed in Nogami et al!

Furthermore, in the Office Action dated January 14, 2009, the Office newly explains the position taken in both the Advisory Action dated December 17, 2008, and the Final Office Action dated July 22, 2008; namely that the Office believes the "103(a) rejection may be overcome by showing the improvement in the physical properties of the coated films prepared according to the instant invention and those prepared as taught in Nogami et al." (Office Action dated January 14, 2009, page 3, lines 6-8). With respect to such "physical properties of the coated films", Applicants submit the following. As disclosed at the bottom of column 2 of Nogami et al, the reference describes the one step preparation of a polysiloxane by the reaction of a tetraalkoxysilane with the alkoxylated silicon compound (B) of formula (2) in a reactive medium that also contains an alkyl alcohol and oxalic acid and possibly a reaction modifier. In application, the mixture may be applied to a substrate and then heat-cured. As stated in the brief abstract attached to the brief, the polycondensation product of the system of Nogami et al gives product films of low refractive index. On the other hand, the hydrolysis of materials such as taught in Hayashi et al results in films of high index of refraction. Thus, in Nogami et al only a single reaction, which is a heat-curing reaction, is conducted, thereby leaving an insoluble coated film on the substrate that has a low refractive index and is water repellant (column 3, lines 27-32). No description anywhere

is found in the Nogami et al reference of a first stage of conducting hydrolysis of a mixture of alkoxylated silicon compounds followed by condensation of certain reaction components to prepare a silica condensate. On this basis alone it can not be said of the two patent disclosures that the reactants and process conditions disclosed therein lead to the formation to the same or similar product.

A further understanding of how different the two processes are of Nogami et al and Hayashi et al can be obtained by the fact that in Nagomi et al the tetraalkoxysilane reactant reacts with a perfluoroalkyl group containing trialkoxysilane when the disclosed anhydrous reaction mixture, also containing oxalic acid and an alcohol, is heat-cured. (The content of any silica in the polysiloxane material is minimal.) No precursor silica based material is prepared, which is subsequently reacted in another reaction step as disclosed in Hayashi et al. The result obtained in Nogami et al is a polysiloxane coating or film that is hardened and water repellant. In Hayashi et al, on the other hand, after having formed component (A) by hydrolysis and condensation reactions as indicated above, component (A), which is largely silica, is reacted with one of the nine compounds which constitute component (B), that are described at column 11, lines 4-12 of the text. None of the compounds which are component (B) compounds contain silicon or are silane compounds.

Hayashi et al also discloses at the top of column 14 that the organic solvent solution of the two components (A) and (B) may contain a silane coupling agent as one of several optional additives to couple reactive species together. Aminopropyltrimethoxysilane (column 15, line 5) is taught by Hayashi et al, as a silane coupling agent, as well as ureidopropyltrimethoxy(triethoxy)silane, (column 15, lines 11 and 12). However, neither the disclosure of Nagomi et al nor the present invention requires a silane coupling agent. Nogami et al does speak to the possible use of aminopropyltrimethoxysilane in the composition of the

reference, but as a modifier (E) which lowers the temperature necessary to achieve curing of the product, not as a silane coupling agent.

It is clear that the two compounds of an aminopropyltrimethoxysilane and ureidopropyltrimethoxy(triethoxy)silane are not functional equivalents. Moreover, in the present invention it is the ureidoalkoxysilane compound that critically distinguishes the present invention over the closest of the prior art references which is the Nagomi et al patent, and is thereby not functionally equivalent to the silane coupling agent of Hayashi et al nor the modifier (E) of Nagomi et al. The present invention claims a composition of a tetraalkoxysilane, a perfluoro group containing silicon compound (B) of formula (2) and an alcohol, and in addition oxalic acid. In the present invention the ureidoalkoxysilane compound (C) functions in a selective manner in that a coated film obtained from the composition maintains a low refractive index *while significantly improving the hardness of the coated film*. This is clear from the comparative evidence presented in the "Refractive Index" and "Abrasion Resistance" columns of Tables 1 and 3 of the present specification. (Coating fluids L_1 - L_5 are within the scope of the present invention, while coating fluid L_6 is outside the scope of the present invention, because of the lack of the presence of a ureidoalkyltrialkoxysilane component in the coating composition.) On the other hand, the Nogami et al patent does not suggest a composition anywhere that results in coated films of significantly improved hardness as a result of the incorporation of a particular additive into the initially prepared composition, specifically a ureidoalkyltrialkoxysilane.

As noted above Nagomi et al teaches the additional possible presence of a modifier (E) and examples of such are disclosed in the paragraph bridging columns 4 and 5 of the patent. These compounds are largely trialkoxysilanes. Aminopropyltriethoxysilane is included as a silicon compound. However, the presence of such a modifier in the composition of the patent as a curing temperature lowering agent results in a distinctly different effect

from the hardening effect achieved by the ureidoalkoxysilane component in the present composition.

The Examiner in the Office Action of January 14, 2009 refers in particular to Example 3 of the Nagomi et al patent as significant. However, the example discloses nothing beyond a typical embodiment of the composition of Nogami et al of an ethanolic solution of tetraethoxysilane, tridecafluorooctyltrimethoxysilane and a modifier combination of glycidyloxypropyltrimethoxysilane and aminopropyltriethoxysilane, along with oxalic acid. There is, of course, no teaching or suggestion of a combination of a tetraalkoxysilane, a perfluoro group containing alkoxysilane compound, a ureidoalkoxysilane in an alcoholic solution also containing oxalic acid.

The Examiner holds at the bottom of page 4 of his remarks that *Nogami et al and Hayashi et al are combinable because they are from the same field of endeavor, namely, polysiloxane films prepared via hydrolysis and condensation of alkoxysilane precursors.* Appellants maintain that this statement is manifestly incorrect. Nogami et al does, in fact, disclose polysiloxane films that are prepared from the tetraalkoxysilane/ perfluoro group containing alkoxysilane ethanolic solution described above. This alcoholic solution is applied to a substrate, and then in one step is cured to a hardened film. On the other hand, as seen from the discussion above concerning Hayashi et al, a first step is conducted in which a product of hydrolysis and condensation is prepared from at least one of the three reactants identified as compounds (1), (2) and (3). The hydrolyzate/condensate formed is largely silica which is characterized substantially of Si-O-Si bonds. This product is not at all produced by the method disclosed in Nogami et al. In fact, Nogami et al discloses the specific limitation at column 2, lines 61-63 that the reaction mixture disclosed therein contains from 0.5 to 10 wt % of silica calculated from the silicon atoms present in the reaction mixture. After the preparation of the hydrolyzate/condensate product of Hayashi et al, the product is reacted

with the ingredient (B) in an organic solvent. There is no such counterpart step in Nogami et al. Ingredient (B) is selected largely from a series of different organic molecules (columns 11-13). Accordingly, it is not correct that both of the cited and applied references teach polysiloxane films prepared by hydrolysis and condensation.

In view of the comments as stated above, appellants continue to believe that the decision by the Examiner to continue the rejection of the claims of the application is erroneous and should be REVERSED.

Respectfully submitted,

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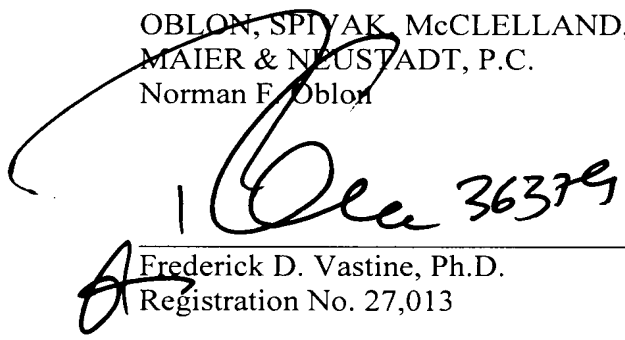

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CLAIMS APPENDIX

Claim 1. A process for preparing a coating fluid containing a polysiloxane, which comprises:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer of from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer of from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group, and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 ,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol and the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.2 to 2 mol, and

heating this reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining at a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water.

Claim 2. The process for preparing a coating fluid according to Claim 1, wherein in the formation of the reaction mixture, in addition to the silicon compounds (A), (B) and (C), the alcohol (D) and the oxalic acid (E), at least one alkylalkoxysilane selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, pentyltrimethoxysilane, pentyltriethoxysilane, heptyltrimethoxysilane, heptyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, dodecyltrimethoxysilane, dodecyltriethoxysilane, hexadecyltrimethoxysilane, hexadecyltriethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, phenyltrimethoxysilane, phenyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, γ-aminopropyltrimethoxysilane, γ-aminopropyltriethoxysilane, γ-glycidoxypropyltrimethoxysilane, γ-glycidoxypropyltriethoxysilane, γ-methacryloxypropyltrimethoxysilane, γ-methacryloxypropyltriethoxysilane, dimethyldimethoxysilane and dimethyldiethoxysilane, is incorporated as a modifier (F) in a ratio ranging from 0.02 to 0.2 mol per mol of the silicon compound (A).

Claim 3. The process for preparing a coating fluid according to Claim 1, wherein at least one sol selected from the group consisting of silica sol, alumina sol, titania sol, zirconia sol, magnesium fluoride sol and ceria sol is incorporated as an additive (G) to the coating fluid.

Claim 4. A process for forming a coating film, which comprises:

forming a reaction mixture comprising a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer of from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group, and

oxalic acid (E), wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups present in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol, and

(iv) the ratio of the oxalic acid (E) per mol of the total alkoxy groups present in the silicon compounds (A), (B) and (C) ranges from 0.2 to 2 mol;

heating this reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture becomes at most 5 mol %, while maintaining at a SiO_2 concentration of from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water, and

forming a solution of a polysiloxane; then

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating; and

heat-curing the coating at a temperature of from 40 to 450° C, to form a coating film having a refractive index of from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, as adhered to the substrate surface.

Claim 5. The process for forming a coating film according to Claim 4, wherein in the formation of the reaction mixture, in addition to the silicon compounds (A), (B) and (C), the alcohol (D) and the oxalic acid (E), at least one alkylalkoxysilane selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, pentyltrimethoxysilane, pentyltriethoxysilane, heptyltrimethoxysilane, heptyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, dodecyltrimethoxysilane,

dodecyltriethoxysilane, hexadecyltrimethoxysilane, hexadecyltriethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, phenyltrimethoxysilane, phenyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, γ-aminopropyltrimethoxysilane, γ-aminopropyltriethoxysilane, γ-glycidoxypropyltrimethoxysilane, γ-glycidoxypropyltriethoxysilane, γ-methacryloxypropyltrimethoxysilane, γ-methacryloxypropyltriethoxysilane, dimethyldimethoxysilane and dimethyldiethoxysilane, is incorporated as a modifier (F) in a ratio of from 0.02 to 0.2 mol per mol of the silicon compound (A).

Claim 6. The process for forming a coating film according to Claim 4, wherein at least one sol selected from the group consisting of silica sol, alumina sol, titania sol, zirconia sol, magnesium fluoride sol and ceria sol is incorporated as an additive (G) to the coating fluid.

Claim 16. The process for forming a coating film according to Claim 4, wherein said SiO_2 concentration ranges from 1 to 8 wt %.

Claim 17. A coating film having a refractive index ranging from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, which is formed as adhered to a substrate surface by

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer of from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups present in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol and the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from of 0.2 to 2 mol;

heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining at a SiO_2 concentration ranging from 0.5 to 10 wt, as calculated from silicon atoms in the reaction mixture and while in the absence of water and forming a solution of a polysiloxane; and

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating; and

heat-curing the coating at a temperature ranging from 40 to 450° C.

Claim 18. The coating film according to Claim 17, wherein in the formation of the reaction mixture, in addition to the silicon compounds (A), (B) and (C), the alcohol (D) and the oxalic acid (E), at least one alkylalkoxysilane selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, pentyltrimethoxysilane, pentyltriethoxysilane, heptyltrimethoxysilane, heptyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, dodecyltrimethoxysilane, dodecyltriethoxysilane, hexadecyltrimethoxysilane, hexadecyltriethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, phenyltrimethoxysilane, phenyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, γ-aminopropyltrimethoxysilane, γ-aminopropyltriethoxysilane, γ-glycidoxypropyltrimethoxysilane, γ-glycidoxypropyltriethoxysilane, γ-methacryloxypropyltrimethoxysilane, γ-methacryloxypropyltriethoxysilane, dimethyldimethoxysilane and dimethyldiethoxysilane, is incorporated as a modifier (F) in a ratio ranging from 0.02 to 0.2 mol per mol of the silicon compound (A).

Claim 19. The coating film according to Claim 17, wherein at least one sol selected from the group consisting of silica sol, alumina sol, titania sol, zirconia sol, magnesium fluoride sol and ceria sol is incorporated as an additive (G) to the coating fluid.

Claim 20. A process for forming a coating film, which comprises:

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer ranging from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer ranging from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group is optionally substituted by one or more substituents of the same or different types selected from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol, and

(iv) the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from 0.2 to 2 mol;

heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water forming a solution of a polysiloxane; and

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating;

drying the coating at a temperature ranging from 40 to 150° C, and

aging the coating at a temperature of from 20 to 100°C for curing, to form a coating film having a refractive index of from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, as adhered to the substrate surface.

Claim 21. The process for forming a coating film according to Claim 20, wherein in the formation of the reaction mixture, in addition to the silicon compounds (A), (B) and (C), the alcohol (D) and the oxalic acid (E), at least one alkylalkoxysilane selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, pentyltrimethoxysilane, pentyltriethoxysilane, heptyltrimethoxysilane, heptyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, dodecyltrimethoxysilane, dodecyltriethoxysilane, hexadecyltrimethoxysilane, hexadecyltriethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, phenyltrimethoxysilane, phenyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, γ-aminopropyltrimethoxysilane, γ-aminopropyltriethoxysilane, γ-glycidoxypropyltrimethoxysilane, γ-glycidoxypropyltriethoxysilane, γ-methacryloxypropyltrimethoxysilane, γ-methacryloxypropyltriethoxysilane, dimethyl-

dimethoxysilane and dimethyldiethoxysilane, is incorporated as a modifier (F) in a ratio of from 0.02 to 0.2 mol per mol of the silicon compound (A).

Claim 22. The process for forming a coating film according to Claim 20, wherein at least one sol selected from the group consisting of silica sol, alumina sol, titania sol, zirconia sol, magnesium fluoride sol and ceria sol is incorporated as an additive (G) to the coating fluid.

Claim 23. A coating film having a refractive index ranging from 1.28 to 1.41 and a contact angle with water ranging from 90° to 115°, which is formed as adhered to a substrate surface by

forming a reaction mixture comprising

a silicon compound (A) of formula (1):

$$Si(OR)_4 \qquad (1)$$

wherein R is a C_{1-5} alkyl group,

a silicon compound (B) of formula (2):

$$CF_3(CF_2)_nCH_2CH_2Si(OR^1)_3 \qquad (2)$$

wherein R^1 is a C_{1-5} alkyl group, and n is an integer of from 0 to 12,

a silicon compound (C) of formula (3):

$$H_2NCONH(CH_2)_mSi(OR^2)_3 \qquad (3)$$

wherein R^2 is a C_{1-5} alkyl group, and m is an integer of from 1 to 5,

an alcohol (D) of formula (4):

$$R^3CH_2OH \qquad (4)$$

wherein R^3 is a hydrogen atom or a C_{1-12} alkyl group, wherein the alkyl group may optionally be substituted by one or more substituents of the same or different types selected

from the group consisting of a C_{1-3} alkyl group, a C_{1-3} hydroxyalkyl group, a C_{2-6} alkoxyalkyl group, a C_{2-6} hydroxyalkoxyalkyl group and a C_{3-6} alkoxyalkoxyalkyl group), and

oxalic acid (E),

wherein

(i) the ratio of the silicon compound (B) per mol of the silicon compound (A) ranges from 0.05 to 0.43 mol,

(ii) the ratio of the silicon compound (C) per mol of the silicon compound (A) ranges from 0.01 to 0.20 mol,

(iii) the ratio of the alcohol (D) per mol of the total alkoxy groups present in the silicon compounds (A), (B) and (C) ranges from 0.5 to 100 mol, and

(iv) the ratio of the oxalic acid (E) per mol of the total alkoxy groups contained in the silicon compounds (A), (B) and (C) ranges from of 0.2 to 2 mol; heating the reaction mixture at a temperature ranging from 40 to 180° C until the total amount of the silicon compounds (A), (B) and (C) remaining in the reaction mixture reaches at most 5 mol %, while maintaining a SiO_2 concentration ranging from 0.5 to 10 wt % as calculated from silicon atoms in the reaction mixture and in the absence of water forming a solution of a polysiloxane;

applying a coating fluid comprising the polysiloxane solution on a substrate surface to form a coating;

drying the coating at a temperature ranging from 40 to 150° C and aging the coating at a temperature ranging from 20 to 100° C for curing.

Claim 24. The coating film according to Claim 23, wherein in the formation of the reaction mixture, in addition to the silicon compounds (A), (B) and (C), the alcohol (D) and the oxalic acid (E), at least one alkylalkoxysilane selected from the group consisting of methyltrimethoxysilane, methyltriethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, pentyltrimethoxysilane, pentyltriethoxysilane, heptyltrimethoxysilane, heptyltriethoxysilane, octyltrimethoxysilane, octyltriethoxysilane, dodecyltrimethoxysilane, dodecyltriethoxysilane, hexadecyltrimethoxysilane, hexadecyltriethoxysilane, octadecyltrimethoxysilane, octadecyltriethoxysilane, phenyltrimethoxysilane, phenyltriethoxysilane, vinyltrimethoxysilane, vinyltriethoxysilane, γ-aminopropyltrimethoxysilane, γ-aminopropyltriethoxysilane, γ-glycidoxypropyltrimethoxysilane, γ-glycidoxypropyltriethoxysilane, γ-methacryloxypropyltrimethoxysilane, γ-methacryloxypropyltriethoxysilane, dimethyldimethoxysilane and dimethyldiethoxysilane, is incorporated as a modifier (F) in a ratio ranging from 0.02 to 0.2 mol per mol of the silicon compound (A).

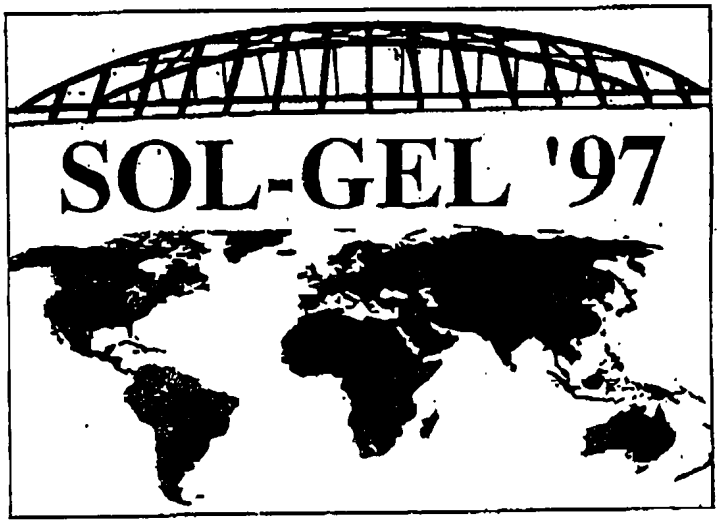
Claim 25. The coating film according to Claim 23, wherein at least one sol selected from the group consisting of silica sol, alumina sol, titania sol, zirconia sol, magnesium fluoride sol and ceria sol is incorporated as an additive (G) to the coating fluid.

EVIDENCE APPENDIX

Appellants enclose a copy of the brief abstract referred to in their discussion of the Nogami et al patent. The abstract is of paper given at the 9th International Workshop of Glasses, Ceramics, Hybrids and Nanocomposites from Gels (1997).

RELATED PROCEEDINGS APPENDIX

No copy of a decision rendered by a court or the Board in any proceeding identified pursuant to paragraph (c)(1)(ii) is enclosed.



ABSTRACTS

9th International Workshop on Glasses, Ceramics, Hybrids and Nanocomposites from Gels

31 August - 5 September 1997

Sheffield, UK

FP33

APPLICATION OF SOL-GEL FILMS FROM TEOS-FLUOROALKYLSILANE-CARBOXYLIC ACID

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Sol-gel films with low refractive index were prepared by polycondensation reactions of TEOS-fluoroalkyisilane-carboxylic acid systems without addition of water. On the other hand, hydrolysis of their materials gave higher refractive index films. For example, refractive index value of film without hydrolysis was 1·38 at curing temperature of 100°C, and that of film from hydrolysis was 1·45. It was considerated that the difference of refractive index property was caused by polymer conformation i.e., the former were linear-like polymer, the latter were cross-linked polymer. In this investigation, we have developed the low refractive index films. These films are very useful to anti-reflection films for LCD's etc.